

# Correlation-enhanced Friedel oscillations in amorphous alloys and quasicrystals

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We show that quantum correlations induced by electron–electron interactions in the presence of random impurity scattering can play an important role in the thermal stabilization of amorphous Hume–Rothery systems: When there is strong backscattering off local, concentric ion clusters, the static electron density response  $\chi(0, q)$  acquires a powerlaw divergence at  $q = 2k_F$  even at elevated temperature. This leads to an enhancement as well as to a systematical phase shift of the Friedel oscillations, both consistent with experiments. The possible importance of this effect in icosahedral quasicrystals is discussed.

## I. INTRODUCTION

A large class of noble–polyvalent metal alloys exhibit a crystalline to amorphous transformation (CAT) as a function of the polyvalent metal content. There are several experimental indications for the amorphous phase being stabilized by the Hume–Rothery (HR) mechanism, i.e. by forming a structure-induced pseudogap in the electronic density of states (DOS) at the Fermi level  $\varepsilon_F$ . The experimental evidence [1] includes the observation of a pronounced pseudogap, a maximum of the electrical resistivity at the CAT, and the coincidence of the atomic spacing  $a$  with the Friedel wave length  $\lambda_F = \pi/k_F$  near the CAT, where  $\hbar k_F$  is the Fermi momentum. Very similar behavior is found in icosahedral (*i*) quasicrystals [2,3]. The conjecture of a HR-like stabilization mechanism is strongly supported by detailed theoretical studies both for amorphous [4] and for quasicrystalline [5–8] systems, although there is also the possibility of entropic stabilization [9]. The fact that in the amorphous state the above-mentioned structural matching is observed over distances of up to  $5\lambda_F$  has lead to the assumption that these remarkably long-range correlations are induced by the ions being bound in the minima of the potential formed by the Friedel oscillations (FO) around an arbitrary central ion.

However, several experimental puzzles have remained unexplained: (1) At finite temperature  $T$  and also in the presence of disorder the impurity-averaged FOs are exponentially damped due to the spread of the Fermi momentum over a width given by  $T$  and the inverse elastic mean free path, respectively. Hence, the stability of amorphous alloys at elevated  $T$ , in particular their intermediate-range structural correlations, are difficult to explain by the *conventional* FOs. (2) In all HR-systems, invariably the amorphous state is thermally most stable just at the CAT [1]. (3) In the amorphous state the ionic positions are systematically shifted compared to the minima of the Friedel potential of a free electron sea [1]. The experimental findings (1)–(3) raise the question of a systematical, composition dependent enhancement and phase shift  $\varphi$  of the FOs, where  $\varphi$  varies from  $\varphi \simeq \pi$  deep inside the amorphous phase to  $\varphi = \pi/2$  at the CAT. As shown below, these problems can be explained by one single quantum effect.

## II. THEORY

In amorphous alloys the electronic motion is diffusive instead of ballistic. Since diffusion, as a dissipative process, is difficult to include in an *ab initio* calculation, we here choose a Feynman diagram technique, where diffusion arises in the formalism by averaging over all random configurations of the system. The Nagel–Tauc condition  $2k_F \simeq 2\pi/a \equiv k_p$  implies strong electronic backscattering off local ion clusters. It has been shown [10–12] that this not only generates a pseudogap but at the same time leads to a substantial enhancement of the electron transport or density relaxation rate  $\tau^{-1}$  over the quasiparticle decay rate  $\tau_{qp}^{-1}$ . This is evidenced experimentally by the anomalously small electrical conductivity,  $\sigma = ne^2\tau/m^*$ . Thus, we have as a generic feature of the amorphous state:  $\tau^{-1} \gg \tau_{qp}^{-1}$ .

We now turn to the calculation of the electron density distribution  $\rho(r)$  around an ion embedded in the disordered electron sea. One does not expect a single ion to generate a sizable phase shift of the FOs at distances  $r > a$ ,  $\rho(r) \propto \cos(2k_F r - \varphi)/r^3$ , since it is equal to the electron backscattering phase off that ion, which is small unless  $\varepsilon_F$  is close to an internal resonance. Instead we consider quantum effects due to disorder and Coulomb interaction and assume, for simplicity, a point-like ion charge and a quadratic band  $\varepsilon_k = \hbar^2 k^2 / 2m^*$ . In a diffusive electron system screening is inhibited, so that the effective Coulomb interaction  $v_q^{eff}(z, Z)$  between electrons with complex frequencies  $z$  and  $z + Z$  acquires a long-range, retarded part [13],

$$v_q^{eff}(z, Z) = \frac{v_q}{\epsilon^{RPA}(Z, q)} \Gamma^2(z, Z, q), \quad v_q = \frac{4\pi e^2}{q^2}, \quad (1)$$

where  $\epsilon^{RPA}(Z, q) = 1 + 2\pi i \sigma / (Z \operatorname{sgn} Z'' + i q^2 D)$  is the disordered RPA dynamical dielectric function and the diffusion vertex, defined in Fig. 1 a), is

$$\Gamma(z, Z, q) = \begin{cases} \frac{i/\tau \operatorname{sgn} Z''}{Z + i q^2 D \operatorname{sgn} Z''} & z''(z + Z)'' < 0 \\ 1 & \text{otherwise.} \end{cases} \quad (2)$$

$D = 1/3 v_F^2 \tau$  and  $''$  denote the diffusion constant and

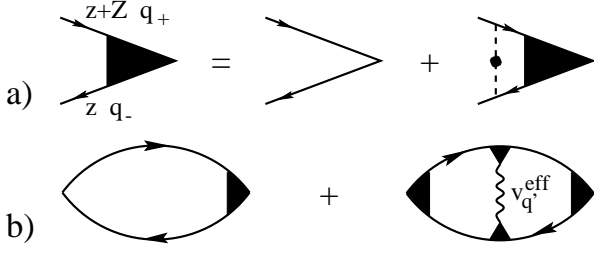


FIG. 1. a) Diffusion vertex  $\Gamma$ . b) Polarisation  $\Pi(0, q)$  including leading order quantum correction induced by disorder and interactions. Dashed lines denote electron-ion scattering, the wavy line with solid triangles the effective Coulomb interaction.

the imaginary part, respectively. The long-range nature of  $v_q^{eff}$  originates from the diffusion pole of  $\Gamma$ . In order to calculate its effect on the FOs, one must consider contributions to the polarisation  $\Pi(0, q)$  where  $\Gamma(z, Z, q')$  enters at  $Z, q' \simeq 0$ , although  $\Pi(0, q)$  is evaluated at large external momenta  $q \simeq 2k_F$ . The leading singular contribution arises from the quantum correction shown in the second diagram of Fig. 1 b). For amorphous metals ( $\tau^{-1} \gg \tau_{qp}^{-1}$ , see above) it is evaluated as [12],

$$\Pi^{(1)}(0, q) = C(\varepsilon_F \tau) \int_{-\varepsilon_F}^{\varepsilon_F} d\nu \frac{1/(4T) \operatorname{sgn}(x-1)}{\cosh^2 \frac{\nu}{2T} \sqrt{|x-1|}}, \quad (3)$$

where  $x = x(\nu) = (q/2k_F)/\sqrt{1+\nu/\varepsilon_F}$  and  $C(\varepsilon_F \tau) = -0.343 [2m^*k_F/(2\pi\hbar)^2]/(\varepsilon_F \tau)^{7/2}$ . The first term of Fig. 1 b),  $\Pi^{(0)}(0, q)$ , corresponds to the Lindhard function, where  $\Gamma$  contributes only a nonsingular factor of  $\mathcal{O}(1)$ . It is seen from Eq. (3) that for  $T = 0$ ,  $\Pi^{(1)}(0, q)$  exhibits a powerlaw divergence  $\propto -\operatorname{sgn}(q-2k_F)/|q-2k_F|^{1/2}$  at  $q = 2k_F$ . Although at finite  $T$  or  $\tau_{qp}^{-1}$  the divergence of  $\Pi^{(1)}(0, q)$  is reduced to a peak, the inverse dielectric function  $1/\varepsilon(q) = 1/(1 - v_q \Pi(0, q))$  still has a  $q = 2k_F$  divergence at a critical transport rate  $\tau_c^{-1}(T)$  even for non-zero single-particle relaxation rate  $\tau_{qp}^{-1} < \tau^{-1}$  and at finite  $T$ . The parameter  $\tau^{-1}$  is varied experimentally by changing the composition of the alloy.

### III. DISCUSSION AND COMPARISON WITH EXPERIMENTS

Fourier transforming  $1 - 1/\varepsilon(q)$  to obtain  $\rho(r)$  [10] shows that for incomplete Fermi surface-Jones zone matching, i.e. small  $\tau^{-1}$ , the quantum corrections generate density oscillations  $\rho^{(1)}(r) \propto -\cos(2k_F r)/r^3$ , which overcompensate the conventional FOs, implying a phase shift of  $\varphi = \pi$  [12]. As  $\tau^{-1} \rightarrow \tau_c^{-1}$ , the increasing  $2k_F$  peak of  $1/\varepsilon(q)$  leads in addition to density oscillations  $\rho^{(1)}(r) \propto \sin(2k_F r)/r^2$ , so that in the vicinity of  $\tau_c^{-1}$

$$\rho(r) \propto -\frac{\cos(2k_F r)}{(2k_F r)^3} + A(\tau^{-1}) \frac{\sin(2k_F r)}{(2k_F r)^2}, \quad (4)$$

with  $A(\tau^{-1}) \simeq 0.343\pi(1 - \tau^{-1}/\tau_c^{-1})^{-1/2}$ . The exponent  $1/2$  is characteristic for diffusive behavior. Thus, the FOs are shifted by  $\varphi = \pi - \tan^{-1}[2k_F r A(\tau^{-1})] \simeq \pi/2 + 1/(2k_F r A)$ , i.e. the diverging Friedel amplitude necessarily goes hand in hand with  $\varphi = \pi/2$ . Note that, in contrast to the conventional FOs, this divergence is robust against damping due to finite  $T$  or disorder. The point where the amplitude  $A$  diverges should be identified with the CAT, since at this point the fluctuations of the Friedel potential also become large, allowing the system to find its crystalline ground state. This resolves in a natural way the problems (1)-(3) mentioned in the introduction.

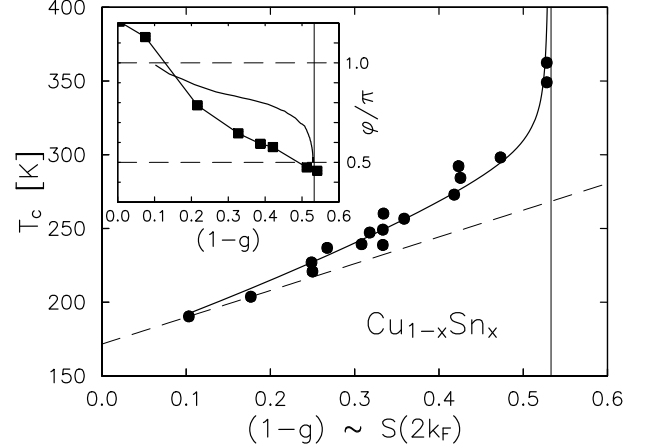


FIG. 2. Crystallization temperature  $T_c$  as a function of the DOS suppression at  $\varepsilon_F$ ,  $(1 - g)$ . Data points represent  $T_c$  for a- $\text{Cu}_{1-x}\text{Sn}_x$  [1]. The solid curve is the fit of the present theory (see text). Vertical line: position of CAT. The inset shows the phase shift  $\varphi$  of the first maximum of the charge density distribution  $\rho(r)$ . Solid line: theory. Data points with solid line: measurements [1] for a- $\text{Cu}_{1-x}\text{Sn}_x$ .

For a direct comparison with experiments the control parameter of the theory,  $\tau^{-1}$ , must be translated into a parameter, which is experimentally accessible: It follows from the scattering theory [11,12] that  $\tau^{-1} = \tau_o^{-1} + \gamma S(2k_F)$ , where the peak of the ionic structure factor  $S(q = 2k_F)$  controls the backscattering amplitude,  $\gamma$  is a constant, and  $\tau_o^{-1}$  is an offset due to momentum independent scattering.  $S(2k_F)$  in turn is proportional [1] to the measured, structure-induced suppression of the DOS  $N(\varepsilon_F)$  at the Fermi level,  $1 - N(\varepsilon_F)/N_o(\varepsilon_F) \equiv 1 - g$ , compared to the free electron gas,  $N_o(\varepsilon_F)$ . The resulting fit of the crystallization temperature  $T_c$  is shown in Fig. 2, where the contribution to the stability coming from the pseudogap formation is assumed to be linear in  $(1 - g)$  (dashed line). The inset shows the calculated phase shift  $\varphi$  and the measured shift of the atomic nearest neighbor position relative to the position of the first conventional Friedel minimum,  $a_o = \pi/k_F$ . Note that there is no adjustable parameter in  $\varphi$ . The general behavior of the shift

is well explained by the theory; however, the experimental data approaches  $\varphi = \pi/2$  faster than predicted. This might be attributed to the fact that, as seen from the discussion after Eq. (4), the higher-order Friedel minima approach  $\varphi = \pi/2$  faster than the first one. In this light, the agreement between theory and experiment is remarkably good.

The structural similarities [3] between amorphous alloys and *i*-quasicrystals suggest that the quantum effect discussed above may be important in the latter systems as well. In fact, quasicrystals seem to fulfill all the necessary preconditions for this effect to occur, i.e. effectively diffusive electron motion [14] and  $\tau^{-1} \gg \tau_{qp}^{-1}$ . The latter is supported by the Fermi surface matching, i.e. by the experimental observation [2] and theoretical prediction [5–7] of structure-induced pseudogaps. Moreover, another more commonly known effect of disorder-enhanced Coulomb interaction, the  $\sqrt{|E - \varepsilon_F|}$  behavior of the DOS in the pseudogap [13], may have been already observed in *i*-quasicrystals by tunneling measurements of the DOS [2]. It is proposed to include the enhanced Friedel potential calculated in the present work in the pseudopotential of more quantitative *ab initio* calculations.

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mun. **30** (1979) 115; B. L. Al'tshuler et al., Phys. Rev. B **22** (1980) 5142.

[14] F. Piéchon, Phys. Rev. Lett. **76** (1996) 4372.

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- [1] For a review and references see P. Häussler, Phys. Rep. **222** (1992) 65.
- [2] D. N. Davydov et al., Phys. Rev. Lett. **77** (1996) 3173; **74** (1995) 3656; Z. M. Stadnik et al. Phys. Rev. Lett. **77** (1996) 1777.
- [3] S. J. Poon, Adv. in Phys. **41** (1992) 303.
- [4] S. S. Jaswal and J. Hafner, Phys. Rev. **B38** (1988) 7311; W. Jank and J. Hafner, Phys. Rev. B **41** (1990) 1497.
- [5] A. P. Smith and N. W. Ashcroft, Phys. Rev. Lett. **59** (1987) 1365.
- [6] T. Fujiwara and T. Yokokawa, Phys. Rev. Lett. **66** (1991) 333.
- [7] J. Hafner and M. Krajčí, Phys. Rev. Lett. **68** (1992) 2321; M. Krajčí, J. Hafner, and M. Mihalcovič, Phys. Rev. B **55** (1997) 843.
- [8] V. E. Dmitrienko and S. B. Astaf'ev, Phys. Rev. Lett. **75** (1995) 1538.
- [9] D. Joseph and V. Elser, Phys. Rev. Lett. **79** (1997) 1066.
- [10] G. D. Mahan, *Many Particle Physics*, (Plenum Press, New York 1990).
- [11] J. Kroha, Physica A **167** (1990) 231.
- [12] J. Kroha, A. Huck, and T. Kopp, Phys. Rev. Lett. **75** (1995) 4278.
- [13] B. L. Al'tshuler and A. G. Aronov, Solid State Com-